

Cyclotron Resonance of Itinerant Holes in Ferromagnetic InMnAs/GaSb Heterostructures

G. A. Khodaparast and J. Kono*

*Department of Electrical and Computer Engineering,
Rice Quantum Institute, and Center for Nanoscale Science and Technology,
Rice University, Houston, Texas 77005*

Y. H. Matsuda,[†] T. Ikaida, S. Ikeda, and N. Miura

Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan

T. Slupinski

Institute of Experimental Physics, Warsaw University, Hoza 69, 00-681 Warsaw, Poland

A. Oiwa and H. Munekata

*Imaging Science and Engineering Laboratory,
Tokyo Institute of Technology, Yokohama, Kanagawa 226-8503, Japan*

(Dated: February 1, 2008)

Abstract

We report the first observation of hole cyclotron resonance (CR) in ferromagnetic InMnAs/GaSb heterostructures both in the high-temperature paramagnetic phase and the low-temperature ferromagnetic phase. We clearly resolve two resonances that exhibit strong temperature dependence in position, linewidth, and intensity. We attribute the two resonances to the so-called fundamental CR transitions expected for delocalized holes in the valence band in the magnetic quantum limit, demonstrating the existence of p -like itinerant holes that are describable within the Luttinger-Kohn effective mass theory.

Keywords:

*Please send all correspondence to: Prof. Junichiro Kono, Rice University, ECE Dept., MS-366, P.O. Box 1892, Houston, TX 77251-1892, U.S.A. Phone: +1-713-348-2209. E-mail:kono@rice.edu

[†]Present address: Department of Physics, Okayama University, Okayama, Japan.

1. INTRODUCTION

The interaction of free carriers with localized spins plays an important role in a variety of magnetic and many-body phenomena in metals.^{1,2,3} Carriers in the vicinity of a magnetic ion are magnetized, which in turn leads to an indirect exchange interaction between magnetic ions. The discovery of carrier-induced ferromagnetism in magnetic III-V semiconductors^{4,5,6} has not only opened up new device opportunities but also provided a novel material system in which to study the physics of itinerant carriers interacting with localized spins. Various theoretical models have been proposed but the microscopic mechanism is still a matter of controversy.^{7,8,9,10} One of the open questions is the nature of the carriers mediating the exchange interaction between Mn ions, i.e., whether they reside in the impurity band (*d*-like), the delocalized valence bands (*p*-like), or some type of mixed states.

In this paper we describe our observation of hole cyclotron resonance (CR) in ferromagnetic InMnAs/GaSb heterostructures, unambiguously demonstrating the existence of delocalized *p*-like carriers. In addition, to our knowledge, this is the first study of CR in any ferromagnetic system covering temperature ranges both below and above the Curie temperature (T_c).^{11,12} CR is a direct and accurate method for determining the effective masses of carriers (i.e., the curvature of the energy dispersion) and therefore the nature of the carrier states. In all the samples studied, we observed two pronounced resonances. Both lines exhibited unusual temperature dependence in their position, intensity, and width. The lower-field resonance showed an abrupt reduction in linewidth with a concomitant decrease in resonance magnetic field slightly above T_c . The higher-field line, which was absent at room temperature, suddenly appeared above T_c , rapidly grew in intensity with decreasing temperature, and became comparable to the lower-field resonance at low temperatures. We ascribe these lines to the two fundamental CR transitions expected for delocalized holes in the valence band of a Zinc-Blende semiconductor in the magnetic quantum limit. We take this as evidence for the existence of a large density of delocalized *p*-like holes in these ferromagnetic systems.

2. SAMPLES STUDIED

Three samples were studied. They were InMnAs/GaSb single heterostructures, consisting of 9-30 nm of InMnAs with Mn content $\sim 9\%$ and a 600-800 nm thick GaSb buffer grown on semi-insulating GaAs (100) substrates. They contained high densities ($\sim 10^{19} \text{ cm}^{-3}$) of holes provided

by the Mn acceptors. The samples were grown by low temperature molecular beam epitaxy. The growth conditions have been described previously.¹³ Unlike the paramagnetic *n*-type¹⁴ and *p*-type¹⁵ InMnAs films we studied earlier, the samples in the present work showed ferromagnetism with T_c ranging from 30 K to 55 K. The magnetization easy axis was perpendicular to the epilayer due to the strain-induced structural anisotropy caused by the lattice mismatch between InMnAs and GaSb (InMnAs was under tensile strain). The energy level structure of InMnAs/GaSb is complicated and known to have a 'broken gap' type-II configuration. The hole densities and mobilities ($\sim 10^{19}$ cm⁻³ and ~ 300 cm²/vs, respectively) were estimated from room temperature Hall measurements and the Curie temperatures were determined by magnetization measurements. Sample 1 was annealed at 250 °C after growth, which increased the T_c by ~ 10 K.^{16,17}

3. EXPERIMENTAL TECHNIQUES

We performed infrared (IR) CR measurements using ultrahigh pulsed magnetic fields (≤ 150 Tesla) generated by the single-turn coil technique.¹⁸ The magnetic field, applied along the growth direction, was measured by a pick-up coil around the sample, which was placed inside a continuous flow helium cryostat. We used IR laser beams with wavelengths of 10.6 μm , 10.2 μm , 9.25 μm (CO₂ laser), and 5.527 μm (CO laser). We circularly polarized the IR radiation using a CdS quarter-wave plate. The transmitted radiation through the sample was collected using a fast liquid-nitrogen-cooled HgCdTe photovoltaic detector. A multi-channel digitizer placed in a shielded room recorded the signals from the pick-up coil and the detector. Although the coil breaks in each shot, the sample and pick-up coil remain intact, making it possible to carry out detailed temperature and wavelength dependence studies on the same specimen. Since the transmission signal was recorded during both the up and down sweeps, each resonance was observed twice in a single pulse. This allowed us to check the reproducibility of observed absorption peaks and to make sure that the spectra were free from any slow heating effects.

4. EXPERIMENTAL RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the transmission of the 10.6 μm beam ($\hbar\omega = 0.117$ eV) through sample 1 ($T_c = 55$ K) and sample 2 ($T_c = 30$ K), respectively, at various temperatures as a function of magnetic field. The beam was hole-active circularly polarized. In Fig. 1(a), from room temperature down to about 80 K, a broad resonance feature (labeled 'A') is observed with

almost no change in intensity, position, and width with decreasing temperature. However, with further decreasing temperature, we observe quite abrupt and dramatic changes in the spectra. First, a significant reduction in linewidth and a sudden shift to a lower magnetic field occur simultaneously. Also, it rapidly increases in intensity with decreasing temperature. In addition, a second feature (labeled 'B') suddenly appears around 125 Tesla, which also grows rapidly in intensity with decreasing temperature and saturates, similar to feature A. At low temperatures, both features A and B do not show any shift in position. Essentially the same behavior is seen for sample 2 in Fig. 1(b).

From Lorentzian fits to the CR data, we deduced the values for the cyclotron mass, density, and mobility. The hole cyclotron masses obtained for this wavelength (10.6 μm) for peaks A and B are $0.051m_0$ and $0.12m_0$, respectively, where m_0 is the mass of free electrons in vacuum. The obtained densities and mobilities for feature A are plotted in Figs. 2(a) and 2(c) for samples 1 and 2, respectively, together with the temperature dependence of the magnetization M in (b) and (d). It is interesting to note that the estimated CR mobility at the lowest temperature of our experiments (~ 15 K) is surprisingly high, i.e., $4\text{--}5 \times 10^3 \text{ cm}^2/\text{Vs}$. This kind of high CR mobility is totally incompatible with the low DC mobilities deduced from Hall measurements, which are $\sim 300 \text{ cm}^2/\text{Vs}$ and sometimes even decrease with decreasing temperature. This suggests that in these ferromagnetic semiconductors a DC mobility is not a good quantity for assessing the CR observability condition, i.e., $\omega_c\tau = B\mu > 1$, where $\omega_c = eB/m^*$ is the cyclotron frequency, τ is the scattering time, and μ is the mobility.

Figure 3(a) shows low temperature CR traces for the three samples taken with 10.6 μm radiation. Both features A and B are clearly observed but their intensities and linewidths vary from sample to sample, depending on the density, mobility, and thickness. The observed unusual temperature dependence is not specific to this particular wavelength. Figure 3(b) displays the wavelength dependence of the CR spectra for sample 2. We can see that both lines shift to higher magnetic fields with decreasing wavelength (i.e., increasing photon energy), as expected. Figures 3(c) and 3(d) show data at different temperatures for sample 1 measured with 9.25 μm and 5.52 μm radiation, respectively. The temperature dependence observed at these shorter wavelengths is similar to what was observed at 10.6 μm . All these data confirm the universality of the effects we observed.

The clear observation of CR indicates that there are delocalized holes in these ferromagnetic samples. This is in agreement with our CR measurements on low- T_c p -type InMnAs films in the paramagnetic phase,¹⁵ which showed similar two resonance CR spectra although the resonances were much broader, temperature dependence was weaker, and the resonance positions were slightly

($\sim 10\%$) lower than the present heterostructure samples. We attribute the resonances to the two CR transitions expected in the magnetic quantum limit (the so-called 'fundamental' transitions¹⁹), one being heavy-hole-like ($m_J = -3/2$) and the other light-hole-like ($m_J = -1/2$). The initial states of these transitions are the two lowest ($n = -1$) Landau levels in the valence band, and the corresponding cyclotron masses at $k_z = 0$ are given by $(\gamma_1 \pm \bar{\gamma})^{-1}m_0$ within the spherical approximation based on a 4×4 Luttinger Hamiltonian,¹⁹ where k_z is the wavenumber in the magnetic field direction and γ_1 and $\bar{\gamma} = (\gamma_2 + \gamma_3)/2$ are Luttinger parameters. More detailed calculations based on an eight-band effective mass model including finite k_z effects successfully reproduced these two resonances for pure InAs and paramagnetic InMnAs films.²⁰ We believe that the $\sim 10\%$ mass enhancement in the heterostructure samples compared to the bulk films is due to quantum confinement (layer thickness only ~ 9 nm) plus non-parabolicity.

We anticipate that the experimental findings presented here will stimulate interest in the problem of the cyclotron resonance of itinerant carriers in ferromagnets, and more theoretical studies will be carried out to explain, in particular, the unusual temperature dependence we observed. We currently have no adequate explanation for the abrupt changes in mass, width and intensity of CR. The rapid line narrowing of the lower-field line as well as the sudden appearance of the higher-field line is equally striking. The ferromagnetic order should split the valence bands *even in the absence of a magnetic field*, which should also strongly modify Landau and Zeeman quantization at high fields. It is important to emphasize that the temperature at which the significant spectral changes start to appear (T_c^*) is consistently higher than the Curie temperature (T_c) in all three samples. This fact could be explainable in light of a recent Monte Carlo study by Schliemann *et al.*,²¹ which showed that short-range magnetic order and finite *local* carrier spin polarization are present for temperatures substantially higher than T_c . Any such order should result in modifications in band structure, which in turn modify CR spectra.

5. SUMMARY

We observed the cyclotron resonance of itinerant holes in ferromagnetic InMnAs/GaSb heterostructures both above and below T_c . We observed two pronounced resonances that were strongly temperature dependent in position, width and intensity. We attribute these transitions to the 'fundamental' light hole and heavy hole cyclotron resonance, the observation of which clearly demonstrates that there are delocalized *p*-like holes in InMnAs. This important information on the carrier states should provide new insight into the microscopic mechanism of carrier-induced

ferromagnetism in this family of magnetic semiconductors.

6. ACKNOWLEDGEMENTS

We gratefully acknowledge support from DARPA MDA972-00-1-0034, NSF DMR-0049024, DMR-0134058 (CAREER), and NEDO.

-
- ¹ P. W. Anderson, Phys. Rev. **124**, 41 (1961).
 - ² C. Kittel, *Indirect Exchange Interaction in Metals*, Solid State Physics **22**, 1 (1968).
 - ³ A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1993).
 - ⁴ H. Ohno, H. Munekata, T. Penny, S. von Molnar, and L. L. Chang, Phys. Rev. Lett. **68**, 2664 (1991).
 - ⁵ H. Munekata, A. Zaslavsky, P. Fumagalli, and R. J. Gambino, Appl. Phys. Lett. **63**, 2929 (1993).
 - ⁶ H. Ohno, A. Shen, F. Matsukura, A. Oiwa, A. Endo, S. Katsumoto, and Y. Iye, Appl. Phys. Lett. **69**, 363 (1996).
 - ⁷ T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science **287**, 1019 (2000); T. Dietl, H. Ohno, and F. Matsukura, Phys. Rev. B **63**, 195205 (2001).
 - ⁸ J. Koenig, H. H. Lin, and A. H. MacDonald, Phys. Rev. Lett. **84**, 5628 (2000); T. Jungwirth, J. Koenig, J. Sinova, J. Kucera, and A. H. MacDonald, cond-mat/0201157.
 - ⁹ V. I. Litvinov and V. K. Dugaev, Phys. Rev. Lett. **86**, 5593 (2001).
 - ¹⁰ A. Chattopadhyay, S. Das Sarma, and A. J. Millis, Phys. Rev. Lett. **87**, 227202 (2001).
 - ¹¹ P. Goy and C. C. Grimes, Phys. Rev. **7**, 299 (1973).
 - ¹² A. K. Chin and A. J. Sievers, J. Appl. Phys. **52**, 7380 (1981).
 - ¹³ T. Slupinski, A. Oiwa, S. Yanagi, and H. Munekata, J. Cryst. Growth **237-239**, 1326 (2002).
 - ¹⁴ M. A. Zudov, J. Kono, Y. H. Matsuda, T. Ikaida, N. Miura, H. Munekata, G. D. Sanders, Y. Sun, and C. J. Stanton, cond-mat/0207082; see also Y. H. Matsuda, T. Ikaida, N. Miura, M. A. Zudov, J. Kono, and H. Munekata, Physica E **10**, 219 (2001).
 - ¹⁵ Y. H. Matsuda, T. Ikaida, N. Miura, Y. Hashimoto, S. Katsumoto, J. Kono, M. A. Zudov, and H. Munekata, Proc. of the 10th Int. Conf. on Narrow Gap Semiconductors and Related Small Energy Phenomena, Physics and Applications (The Institute of Pure and Applied Physics, Tokyo, 2001), pp. 93-95.
 - ¹⁶ T. Hayashi, Y. Hashimoto, S. Katsumoto, and Y. Iye, Appl. Phys. Lett. **78**, 1691 (2001).
 - ¹⁷ S. J. Potashnik, K. C. Ku, S. H. Chun, J. J. Berry, N. Samarth, and P. Schiffer, Appl. Phys. Lett. **79**, 1495 (2001).
 - ¹⁸ K. Nakao, F. Herlach, T. Goto, S. Takeyama, T. Sakakibara, and N. Miura, J. Phys. E: Sci. Instrum. **18**, 1018 (1985).

- ¹⁹ See, e.g., B. D. McCombe and R. J. Wagner, in *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press, New York, 1975), p. 59; see also C. R. Pidgeon and R. N. Brown, Phys. Rev. **146**, 575 (1966).
- ²⁰ G. D. Sanders, Y. Sun, C. J. Stanton, G. A. Khodaparast, J. Kono, Y. H. Matsuda, N. Miura, T. Slupinski, A. Oiwa, and H. Munekata, 2nd Int. Conf. on the Phys. and Appl. of Spin Related Phenomena in Semicond., July 2002, Germany (to be published in J. of Superconductivity).
- ²¹ J. Schliemann, J. Koenig, and A. H. MacDonald, Phys. Rev. B **64**, 165201 (2001).

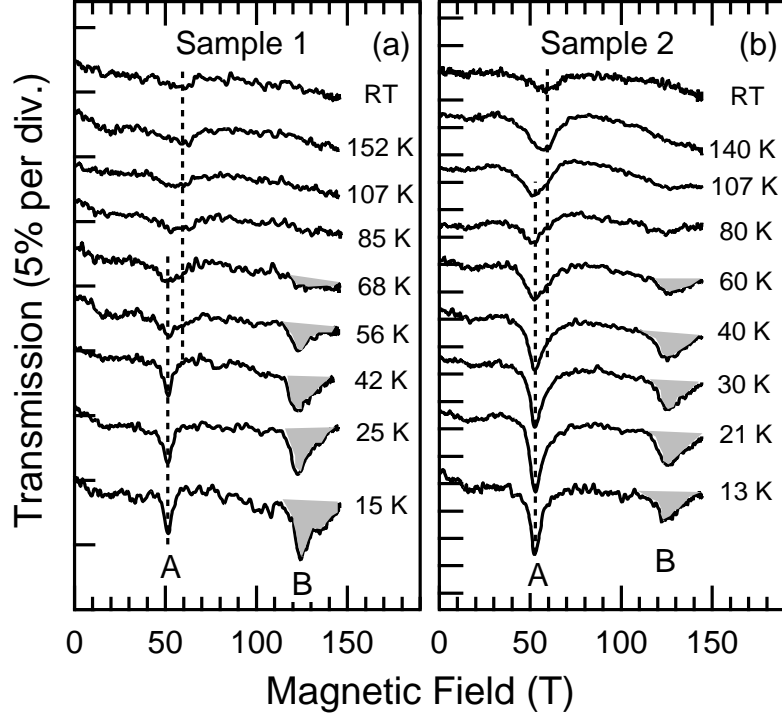


FIG. 1: Cyclotron resonance spectra for (a) sample 1 and (b) sample 2. The transmission of hole-active circularly polarized $10.6 \mu\text{m}$ radiation ($\hbar\omega = 0.117 \text{ eV}$) is plotted as a function of magnetic field at different temperatures. Both samples show two strongly temperature-dependent features, labeled A and B, whose origins are discussed in the text.

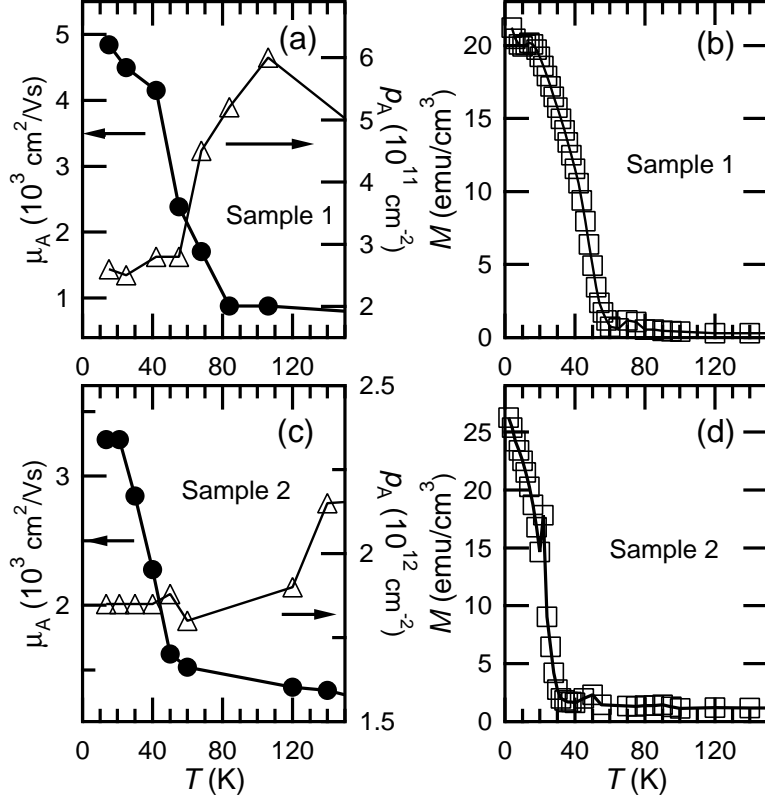


FIG. 2: (a) and (c): Hole mobilities (μ_A) and densities (p_A) vs. temperature (T), deduced from the integrated intensity and linewidth of feature A in Fig. 1. (b) and (d): Magnetization (M) vs. temperature for samples 1 and 2, obtained by SQUID measurements with a magnetic field of 0.5 mT applied along the growth direction.

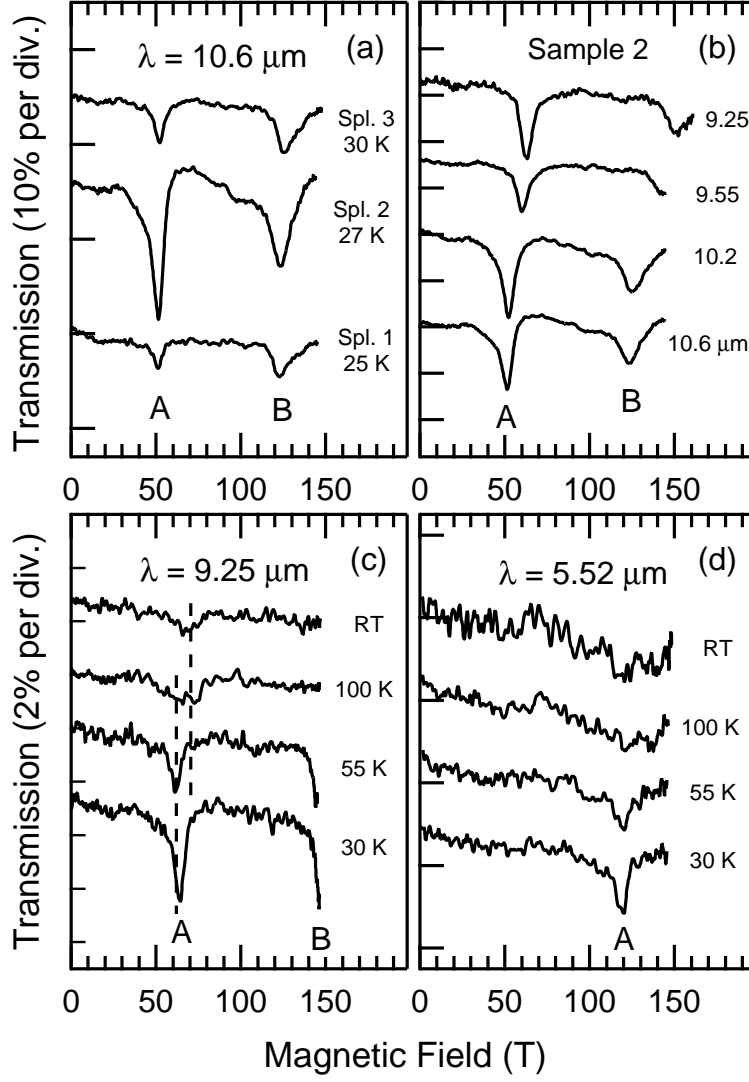


FIG. 3: (a) Low temperature CR spectra for the three samples at $10.6 \mu\text{m}$. (b) Wavelength dependence of the CR spectra for sample 2 at 27 K. (c) CR spectra for sample 1 at different temperatures at $9.25 \mu\text{m}$. (d) CR spectra for sample 1 at $5.52 \mu\text{m}$.